Viscosity Measurements on Colloidal Dispersions (Nanofluids) for Heat Transfer Applications

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Abstract:
This article reports viscosity data on a series of colloidal dispersions collected as part of the International Nanofluid Property Benchmark Exercise (INPBE). Data are reported for seven different fluids that include dispersions of metal-oxide nanoparticles in water, and in synthetic oil. These fluids, which are also referred to as 'nanofluids,' are currently being researched for their potential to function as heat transfer fluids. In a recently published paper from the INPBE study, thermal conductivity data from more than 30 laboratories around the world were reported and analyzed. Here, we examine the influence of particle shape and concentration on the viscosity of these same nanofluids and compare data to predictions from classical theories on suspension rheology.

Zusammenfassung:

In diesem Beitrag werden experimentelle Daten zur Viskosität verschiedener kolloidaler Dispersionen vorgestellt, welche im Rahmen einer internationalen vergleichenden Ringmessung (International Nanofluid Property Benchmark Exercise INPBE) gewonnen wurden. Es werden hierbei sieben unterschiedliche Fluide betrachtet, unter anderem Wasser beziehungsweise synthetisches Oel mit suspendierten metalloxidischen Nanopartikeln. Diese auch Nanofluide genannten Dispersionen werden derzeit intensiv aufgrund ihres moeglichen Potenzials als effizientes Wärmeträger-Fluid untersucht. Vor Kurzem wurden in einem weiteren Beitrag der INPBE-Studie Daten zur Wärmeleitfähigkeit von Nanofluiden veröffentlicht, wie sie in den über 30 teilnehmenden Forschungslabors weltweit gemessen wurden. In diesem Beitrag wird der Einfluss der Partikel-Gestalt und der Partikel-Konzentration auf die Viskosität der gleichen Nanofluide untersucht und mit Korrelationen aus der klassischen Rheologie von Suspensionen verglichen.

Résumé:

Cet article présente des données de viscosité sur des séries de dispersions colloïdales qui font partie du «International Nanofluid Property Benchmark Exercise (INPBE)». Les données correspondent à sept fluides différents incluant des dispersions de nano particules d'oxyde métalliques dans de l'eau et dans de l'huile synthétique. Ces fluides, également connus comme « nano fluides » font actuellement l'objet d'études pour leur potentiel comme des fluides pour le transfert de chaleur. Dans un article récemment publié dans le cadre d'un étude INPBE, des données de conductivité thermique de plus de 30 laboratoires différents ont été présentées et analysées. Dans ce travail, on étudie l'influence de la forme et de la concentration des particules sur la viscosité des nano fluides qui en sont issus. Ces données sont comparées aux prédictions théoriques des modèles classiques sur la rhéologie de suspensions.

Sommario:

Questo articolo riporta dati sperimentali sulla viscosita' di una serie di sistemi di nano-colloidi (nanofluidi) nel contesto del programma internazionale INPBE (International Nanofluid Property Benchmark Exercise). I dati riguardano sette diversi sistemi, comprendenti particelle di ossidi metallici in acqua ed olio sintetico. Questi fluidi sono attualmente soggetti di interesse nella comunita' scientifica per la loro possibile applicazione come fluid termici. I dati di conducibilita' termica prodotti da piu' di 30 laboratori internazionali sono stati riportati e discussi in un articolo su INPBE di recente pubblicazione. Invece qui esaminiamo l'influenza della forma e concentrazione delle nanoparticelle sulla viscosita' degli stessi campioni, e confrontiamo i dati con i risultati delle classiche teorie reologiche per sospensioni colloidali.

Key words: nanofluids, colloidal dispersion, viscosity, thermal conductivity

1 INTRODUCTION

Suspensions of nanometer-sized (1–100 nm), solid particles, also known as colloids or colloidal dispersions, are of great technological importance and can be found in applications ranging from inks to pharmaceuticals. For over a decade, colloidal dispersions containing metallic, or metaloxide, nanoparticles, sometimes referred to as 'nanofluids,' have been the focus of intensive research because of their potential as heat transfer fluids. This research has been driven by the possibility of having fluids with enhanced thermal conductivity, relative to the suspending liquid (basefluid), while mitigating problems associated with erosion, sedimentation and clogging that plague suspensions of larger particles. Interest in nanofluids as heat transfer fluids has been further intensified by early studies reporting anomalously large – relative to classical model predictions – enhancement of thermal conductivity. Several review papers on nanofluid heat transfer have been published [1–4].

Transport properties (e.g. electrical or thermal conductivity) of heterogeneous systems are often estimated using effective medium theory. For example, the effective thermal conductivity k of a suspension composed of solid particles with thermal conductivity k_p at volume fraction ϕ (« 1) dispersed in a liquid of thermal conductivity k_f (« k_p) can be written as

$$\frac{k}{k_f} = 1 + \left[k \right]\phi + O\left(\phi^2\right)$$

(1)

where $[k]$ is defined as follows: $[k] = lim_{\phi \to o}\{(k/k_f - 1)/\phi\}$. Over 100 years ago, Maxwell [5] derived an expression for the thermal (electrical) conductivity of a dilute system of spherical particles that yielded $[k] = 3$. Models for the effective thermal conductivity of heterogeneous systems have been formulated to account for the effects of non-spherical particle shape, particle-particle interactions, and interfacial resistance between the particle and continuous phases [1–4].

There have been numerous experimental studies reporting thermal conductivity data on nanofluids. Much of the reported data show both quantitative and qualitative differences from the effective medium theory prediction in Equation 1. For example, in many early studies, the measured levels of thermal conductivity enhancement for dilute nanofluids ($\phi \leq 0.01$) were significantly larger than the classical prediction given in Equation 1 with $[k] = 3$. Several investigators have reported a dependence of thermal conductivity enhancement on particle size and on temperature, neither of which is consistent with Equation 1. In addition to generating a great deal of optimism about the potential of nanofluids for use as heat transfer fluids, early experimental results also sparked theoretical interest to identify the mechanism(s) responsible for the observed departures from effective medium theory predictions. Further discussion of both experimental and theoretical research on nanofluids can be found elsewhere [1–4]. At present, the study of nanofluids as heat transfer fluids remains both an active and controversial field of research.

It is well known that the addition of solid particles to a liquid can significantly alter its rheological behavior. In shear flows, the apparent viscosity of a fluid η is ratio of the shear stress to the shear rate: $\eta = \sigma/\dot{\gamma}$. The zero-shear rate viscosity is defined as: $\eta_o = lim_{\dot{\gamma} \to o}\{\eta\}$. The zero-shear rate viscosity η_o of a dilute suspension with liquid phase viscosity η_f can be expressed as follows:

$$\frac{\eta_o}{\eta_f} = 1 + \left[\eta\right]\phi + O\left(\phi^2\right)$$

(2)

where the intrinsic viscosity $[\eta]$ of the suspension is defined as: $[\eta] = lim_{\phi \to o}\{(\eta_o/\eta_f - 1)/\phi\}$. The well-known prediction of Einstein [6] for a suspension of spherical particles gives $[\eta] = 2.5$, while for dilute suspensions of particles with large aspect ratio p, $[\eta] \approx p$ [7]. The linear dependence of η_o on particle volume fraction in Equation 2 typically holds for $\phi \leq 0.03$ [7].

In addition to thermal conductivity, it is also evident that viscosity has a significant impact on the overall performance of a heat transfer fluid. Clearly, pumping a fluid with increased viscosity through a heat exchanger requires an increase in pumping energy, thereby reducing the overall benefit of a higher thermal conductivity fluid. In light of this, there have been a number of studies reporting the viscosity of nanofluids [8-14]. These studies considered nanofluids composed of metal-oxide (Al_2O_3, CuO, TiO_2, Fe_2O_3) and metal (Cu), nanometer-sized, spherical particles in water, ethylene glycol, or hydrocarbon oil at concentrations $\phi \leq 0.05$. For all of these nanofluids, Newtonian behavior was observed, and the dependence of relative viscosity η/η_f was approximately linear in particle concentration ϕ. However, the reported intrinsic viscosity $[\eta]$ for these nanofluids was in the range 4-16, which is substantially larger than the theoretical value of 2.5. Prasher et al., [9] and Garg et al., [12] proposed criteria for the overall effectiveness of nanofluids as heat transfer fluids suggesting that $[\eta]$ should be $4 - 5$ times smaller than $[k]$ appearing in Equation 1. The effect of changes in specific heat due to the presence of nanoparticles on the effectiveness of nanofluids as heat transfer fluids has also been investigated [15].

At the first scientific conference centered on nanofluids (Nanofluids: Fundamentals and Applications, September 16-20, 2007, Copper Mountain, Colorado), it was decided to launch an International Nanofluid Property Benchmark Exercise (INPBE), to resolve the inconsistencies in the database and help advance the debate on nanofluid properties. Thermal conductivity data on eight different nanofluids from this study, involving over 30 laboratories around the world, are contained in a recently published paper [16]. A subset of INPBE participants also collected viscosity data on the same set of nanofluids. These data are reported in this paper. The methodology and samples used for the INPBE are described in Section 2. The viscosity data are presented and discussed in Section 3. The findings of this study are summarized in Section 4.

2 INPBE METHODOLOGY

Eight different nanofluids were distributed in four sets to participating INPBE laboratories. To mini-

mize spurious effects due to nanofluid preparation and handling, all organizations were given identical samples from these sets, and were asked to adhere to the same sample handling protocol. The exercise was 'semi-blind,' as only minimal information about the samples was given to the participants at the time of sample shipment. The data were then collected and posted, on the INPBE website (http://mit.edu/nse/nanofluids/benchmark/index.html). Additional details of the INPBE methodology and list of participants can be found in the paper containing the thermal conductivity data [16] and at the INPBE website.

Table 1 gives the main characteristics of the nanofluids used in the INPBE study; the sample names in the first column of the table indicate Set#Sample# as designated in the previous paper [16]. Samples S1S1-S1S7 were provided by Sasol. Sample S1S2 is simply de-ionized H_2O and is the base fluid for S1S1; sample S1S7 is a mixture of poly-alpha olefin (PAO) oil (SpectraSyn-10 by Exxon Oil) and 5%wt. dispersant (Solsperse 21000 by Lubrizol Chemical) and is the base fluid for samples S1S3-S1S6. The nanofluids designated as S1S1 and S1S3-S1S6 contain alumina (Al_2O_3) nanoparticles with the shapes and sizes indicated in Table 1. Sample S3S1 was supplied by W. R. Grace & Co. and is a colloidal dispersion of silica (SiO_2) nanoparticles dispersed in de-ionized H_2O stabilized by the addition of small amounts of Na_2SO_4 (pH = 9). Finally, samples in Set 4 were supplied by Dr. Jorge Gustavo Gutierrez of the University of Puerto Rico – Mayaguez (UPRM). Sample S4S2 is a solution of de-ionized H_2O (75% wt) and tetramethylammonium hydroxide $[(CH_3)_4NOH]$ stabilizer (25% wt). Sample S4S1 contains Mn-Zn ferrite ($Mn_{1/2}$-$Zn_{1/2}$-Fe_2O_4) nanoparticles dispersed in a based fluid S4S2. To summarize, this study involves seven nanofluids and three base fluids. Note that one of the eight nanofluids that were part of the INPBE study, a dilute colloidal gold dispersion, is excluded from this paper because its rheology was indistinguishable from that of water. Additional details concerning these samples and their characterization can be found elsewhere [16].

Viscosity data on each of the fluids listed in Table 1 were reported by approximately 8-10 different laboratories. All reported data were obtained using well-established methods with commercial viscometers that can be 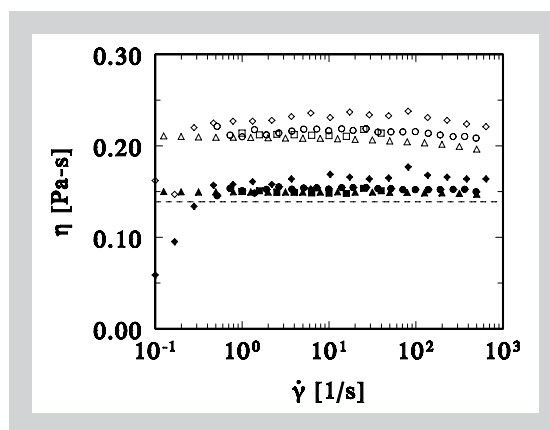broadly separated into two categories according to the type of deformation applied to the sample. In one category, that includes gravity driven capillary, concentric cylinder and immersion viscometers, sample deformation is non-homogeneous and the (average) rate of deformation is usually not known. In the other category are viscometers that apply a controlled and (approximately) uniform deformation to the sample such as cone-plate viscometers. Only two of the 10 samples considered in this study showed non-Newtonian behavior (i.e., viscosity η was dependent on shear rate $\dot{\gamma}$), so that data from both types of viscometers are included for the eight Newtonian fluids. For the two samples displaying non-Newtonian behavior, only data from cone-plate viscometers will be presented. Finally, all data reported here were collected at room temperature, which was in the range 20-26 °C. We recognize that changes in viscosity of roughly 10% are possible over this range of temperatures.

3 RESULTS AND DISCUSSION

First we examine the viscosity versus shear rate behavior of several fluids listed in Table 1. Data for samples S1S3 and S1S4 from four different laboratories are shown in Figure 1. The dashed line in Figure 1 represents the average viscosity of the base fluid, sample S1S7. From this figure, it is clear that these nanofluids are Newtonian over the range of shear rates considered. This is expected given that these fluids contain spherical particles at relatively modest volume fractions (ϕ = 0.01, 0.03); this is consistent with previous results on

Sample	liquid	solid (shape)	d or L·d [nm]	ϕ	η_0 [Pa·s]
S1S1	H_2O	Al_2O_3(rod)	80·10	0.01	$12\pm2\cdot10^{-4}$
S1S2	H_2O	--	--	--	$8.9\cdot10^{-4*}$
S1S3	PAO+surf	Al_2O_3(sph)	100	0.01	$1.7\pm0.4\cdot10^{-1}$
S1S4	PAO+surf	Al_2O_3(sph)	100	0.03	$2.4\pm0.5\cdot10^{-1}$
S1S5	PAO+surf	Al_2O_3(rod)	80·10	0.01	$2.1\pm0.5\cdot10^{-1}$
S1S6	PAO+surf	Al_2O_3(rod)	80·10	0.03	$4.4\pm0.8\cdot10^{-1}$
S1S7	PAO+surf	--	--	--	$1.4\pm0.3\cdot10^{-1}$
S3S1	H_2O+stab	SiO_2(sph)	22	0.32	$4\pm0.8\cdot10^{-1}$
S4S1	H_2O+stab	$Mn_{1/2}Zn_{1/2}Fe_2O_3$(sph)	7	0.0017	$3.11\pm0.37\cdot10^{-3}$
S4S2	H_2O+stab	--	--	--	$3.1\pm0.41\cdot10^{-3}$

similar systems [8 - 14]. It is also evident from Fig-
ure 1 that there is good agreement between the
data reported by the different laboratories with
variations of approximately 10%. Results similar
to those shown in Figure 1 were obtained for sam-
ples S1S1, S1S7, S4S1 and S4S2; therefore only the
average viscosity of these Newtonian fluids will
be presented later in this section.

Figure 2 shows the viscosity versus shear
rate behavior of the concentrated colloidal silica
dispersion sample S3S1. At low shear rates, these
data appear to approach a constant viscosity η_o
with a value of approximately 0.4 Pa-s, and a con-
stant viscosity at shear rates greater than 10 s^{-1}.
At intermediate shear rates, between 0.1 - 10 s^{-1},
pronounced shear thinning is observed. The scat-
ter among the data from different laboratories is
larger in Figure 2 than Figure 1, and is probably
the result of the higher sensitivity to shear his-
tory of the more concentrated system, as
explained next. Shear thinning behavior, which
is typical for concentrated suspensions of spher-
ical particles, can be explained by the flow-
induced perturbation of particle positions [7].
The rate of particle displacement by flow is giv-
en roughly by the shear rate $\dot{\gamma}$, and the rate of
particle movement by Brownian motion is
roughly $k_BT/\eta d^3$. Hence, when flow perturbs par-
ticle positions at a rate faster than they are recov-
ered by thermal fluctuations, the viscosity of the
suspension becomes shear-rate dependent. The

inset to Figure 2 shows a commonly used means
of rescaling viscosity and shear rate to demon-
strate the competition between convection
(flow) and diffusion (Brownian motion) in sus-
pensions [7].

Viscosity versus shear rate data from four
different laboratories for the nanofluids com-
prised of rod-shaped Al$_2$O$_3$ particles in oil (S1S5
and S1S6) are shown in Figure 3. The dashed line
in Figure 3 represents the average viscosity of the
base fluid (S1S7). From this figure, we see that the
lower concentration (ϕ = 0.01) fluid is Newton-
ian, while the higher concentration (ϕ = 0.03) flu-
id is non-Newtonian. Again, agreement between
the different laboratories is good. Comparison of
the data in Figures 1 and 3 for ϕ = 0.03 shows, as
expected, that the viscosity of the fluid with non-
spherical particles is more sensitive to shear com-
pared to the fluid with spherical particles. For
non-spherical particles, non-Newtonian effects
are the result of flow-induced perturbation of
particle orientations from the random state that
exists at equilibrium [7]. In this case, it is a com-
petition between the tendency for flow to orient
particles and thermally driven rotation of parti-
cles.

Viscosity data for the nanofluid comprised
of spherical Al$_2$O$_3$ particles with ϕ = 0.03 (S1S4)
and its oil base fluid (S1S7) as reported by differ-
ent INPBE participants are shown in Figure 4. For
a given laboratory, the reported uncertainty was
typically around ± 5%, which is roughly the size
of the symbols in Figure 4. The data in this figure
give an idea of consistency of viscosity data
reported by different laboratories using different
techniques, which is within approximately ±
20%. Similar results were found for the other
nanofluids used in this study.

The relative viscosity versus particle con-
centration for nanofluids with spherical particles
and rod-shaped particles are shown in Figures 5
and 6 respectively. As expected, the data in both
figures indicate a linear dependence of η/η_f on ϕ.
The measured dependence [η], however, is
roughly ten times larger than predicted, with
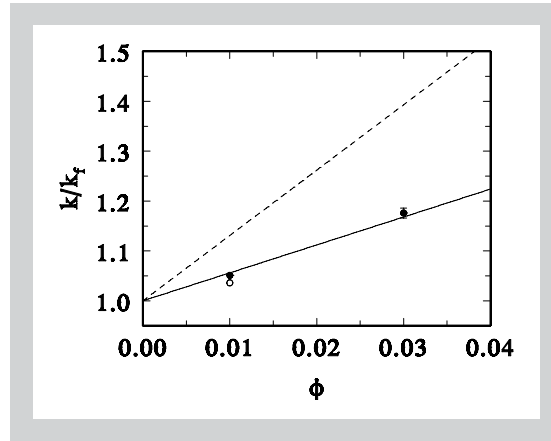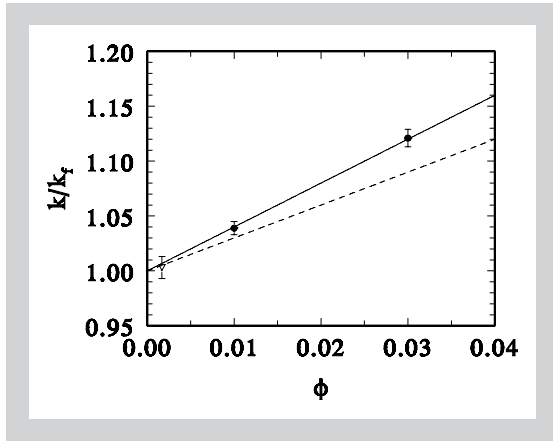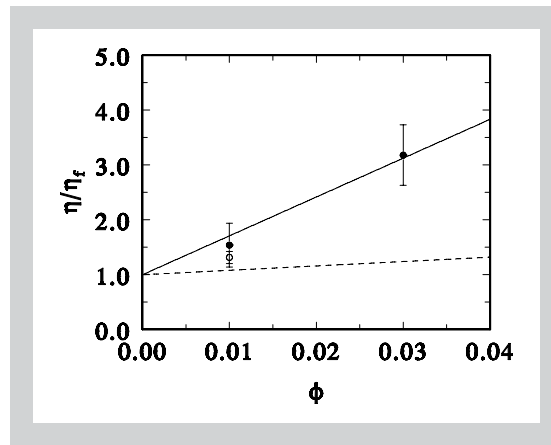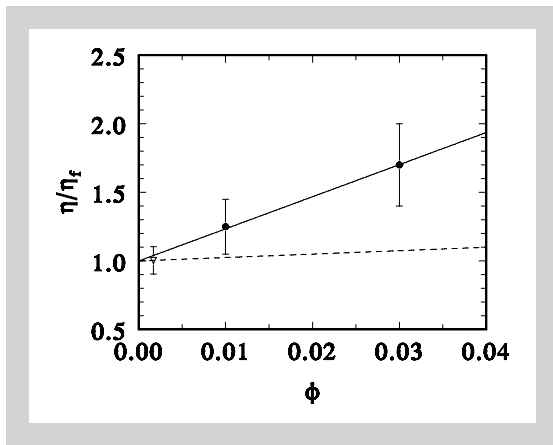
$[\eta] = 23.4$ for the spherical particles and $[\eta] = 70.8$ for the rod-shaped particles. The shear thinning behavior observed in alumina nanorod suspensions can also be due to shear induced reorganization of nanorods under confined geometry where the packing can become more efficient. A similar result has been reported in recently published data for nanofluids with spherical particles [8–13], although somewhat lower values of $[\eta] = 4$–16 were found. A possible explanation for these pronounced differences between theory and experiment is particle agglomeration, which would increase the effective volume fraction of the particles [10–14]. Indeed, light scattering results on these fluids reported elsewhere [16] are consistent with the occurrence of particle agglomeration.

For comparison, we show the relative thermal conductivity versus particle concentration data from the INPBE study [16] in Figures 7 and 8. As with the viscosity data, nanofluids with both spherical and rod-shaped particles show a linear dependence of k/k_f on ϕ. The data for the fluids with spherical particles in Figure 7 show a slightly stronger dependence on particle concentration than predicted: measured $[k] = 4.0$, and predicted $[k] = 3$. This observation is consistent with the presence of particle agglomeration in these systems, and suggests that particle clustering has a larger effect on viscosity than thermal conductivity in nanofluids with spherical particles. The situation

is different for the nanofluids with rod-shaped particles as shown in Figure 8. Here, the observed dependence on particle concentration is weaker than predicted: measured $[k] = 5.6$, and predicted $[k] = 13.1$. Apparently, particle agglomeration can significantly reduce the effective thermal conductivity of nanofluids with rod-shaped particles. A second explanation for the reduction in effective thermal conductivity in these nanofluids is interfacial thermal resistance [16].

4 CONCLUSIONS

Viscosity data have been collected as part of an International Nanofluid Property Benchmark Exercise (INPBE). These data are from approximately 10 different laboratories around the world on a series of 10 different nanofluids and their base fluids. In general, the agreement between different laboratories was good with variations of approximately ± 20%, which, in part, could be explained by lab-to-lab temperature variations. Two of seven nanofluids showed shear-thinning behavior; the remaining five showed Newtonian behavior. For nanofluids with both spherical and rod-shaped nanoparticles, the dependence of viscosity (relative to the base fluid viscosity) on particle concentration (volume fraction) was significantly stronger than predicted by dilute suspension theory. This discrepancy was attributed to particle agglomera-

Map:
Map showing locations of laboratories participating in the International Nanofluid Properties Benchmark Exercise (INPBE).

tion. In contrast, the observed enhancement in thermal conductivity was slightly larger for the spherical particle fluids, and significantly lower for the rod-shaped particle fluids, than predicted by effective medium theory. As noted in the introduction, criteria for the overall effectiveness of nanofluids as heat transfer fluids have been proposed [9, 12], which suggest [η] should be 4 - 5 times smaller than [k]. Clearly, the nanofluids considered in this study would fail; this suggests that the overall effect of adding nanoparticles to the base fluid is negative in terms of heat transfer performance.

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